

Magnon dispersions in quantum Heisenberg ferrimagnetic chains at zero temperature

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Within the Dyson-Maleev boson formalism, we study the zero-temperature magnon dispersions in a family of one-dimensional quantum Heisenberg ferrimagnets composed of two different spins (S_1, S_2) in the elementary cell. It is shown that the spin-wave theory can produce precise quantitative results for the low-energy excitations. The spin-stiffness constant ρ_s and the optical magnon gap Δ of different (S_1, S_2) ferrimagnetic systems are calculated, respectively, to second and third order in the quasiparticle interaction. The spin-wave results are compared with available numerical estimates.

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I. INTRODUCTION

In the last decade a large variety of quasi-one-dimensional (1D) mixed-spin compounds with ferrimagnetic properties has been synthesized.¹ Most of them are molecular magnets containing two different transition-metal magnetic ions which are alternatively distributed on the lattice (Fig. 1). For example, two families of such compounds read $\text{ACu(pba)}(\text{H}_2\text{O})_3 \cdot n\text{H}_2\text{O}$ and $\text{ACu(pbaOH)}(\text{H}_2\text{O})_3 \cdot n\text{H}_2\text{O}$, where pba = 1,3-propylenebis (oxamato), pbaOH = 2-hydroxo-1,3-propylenebis (oxamato), and $\text{A} = \text{Ni, Fe, Co, and Mn}$. Published experimental work implies that the magnetic properties of these mixed-spin materials are basically described by a quantum Heisenberg spin model with antiferromagnetically coupled nearest-neighbor localized spins:

$$\mathcal{H} = J \sum_{n=1}^N [\mathbf{S}_1(n) + \mathbf{S}_1(n+1)] \cdot \mathbf{S}_2(n) - \mu_B H \sum_{n=1}^N [g_1 S_1^z(n) + g_2 S_2^z(n)], \quad J > 0. \quad (1)$$

Here the integers n number the N elementary cells, each of them containing two site spins with quantum numbers $S_1 > S_2$. The g -factors, related to the spins S_1 and S_2 , are denoted by g_1 and g_2 , respectively. μ_B is the Bohr magneton, and H is the external uniform magnetic field applied along the z direction. As an example, the following values for the parameters in Eq. (1) have been extracted from magnetic measurements on the recently synthesized quasi-1D bimetallic compound $\text{NiCu(pba)}(\text{D}_2\text{O})_3 \cdot 2\text{D}_2\text{O}$: $(S_1, S_2) \equiv (S_{\text{Ni}}, S_{\text{Cu}}) = (1, \frac{1}{2})$, $J/k_B = 121\text{K}$, $g_1 \equiv g_{\text{Ni}} = 2.22$, $g_2 \equiv g_{\text{Cu}} = 2.09$.² In view of the recent developments in the physics of uniform quasi-1D systems (concerning, in particular, the behavior of antiferromagnetic chains and ladders in external magnetic fields: see, e.g., Ref. 3 and references therein), ferrimagnetic chains and ladders open an interesting new

area. The point is that the presence of two or more different quantum spins in the elementary cell considerably increases the number of situations of interest: the topology of spin arrangements plays an essential role in the structure of the ground state and low-energy excitations.⁴ For example, the diamond lattice in Fig. 1 represents another interesting class of Heisenberg ferrimagnetic chains constructed of one kind of antiferromagnetically coupled site spins but with a different number of lattice sites in the \mathcal{A} and \mathcal{B} sublattices.⁵ It is remarkable that only in the last few years interest in the physical properties of 1D quantum Heisenberg ferrimagnets has considerably increased:⁶ most of the early efforts were concentrated on the chemistry of molecular magnets and a relatively small amount of work was devoted to physical properties.⁷

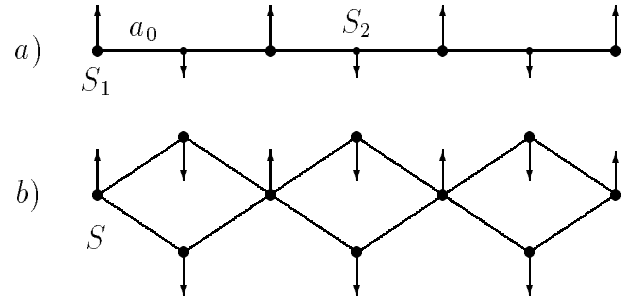


FIG. 1. Two types of ferrimagnetic chains on bipartite lattices: a) mixed-spin chain with alternating site spins S_1 and S_2 ($S_1 > S_2$); b) uniform-spin diamond chain with a site spin S .

Since interactions in the models presented in Fig. 1 set up bipartite lattices, the Lieb-Mattis theorem⁸ is applicable. In particular, for the mixed-spin system it predicts the existence of a ferrimagnetic ground state with the total-spin quantum number $S_g = (S_1 - S_2)N$. Thus the model (1) exhibits long-range ordered magnetic

ground state characterized by the magnetization density $M_0 = (S_1 - S_2)/2a_0$, a_0 being the lattice constant. Such a magnetic phase may be referred to as a *quantized unsaturated ferromagnetic phase*: it is characterized by both the quantized ferromagnetic order parameter \mathbf{M} (quantized in integral or half-integral multiples of the number of elementary cells, $M = M_0N$) and the macroscopic sublattice magnetizations $\mathbf{M}_A = \sum_{n=1}^N \langle \mathbf{S}_1(n) \rangle$ and $\mathbf{M}_B = \sum_{n=1}^N \langle \mathbf{S}_2(n) \rangle$. What makes such a state and the related transitions to magnetically disordered states interesting is the fact that the ferromagnetic order parameter is a conserved quantity: in Heisenberg systems this is expected to imply strong constraints on the critical behavior.⁹ Consequences of the spontaneous symmetry breaking $SO(3) \rightarrow SO(2)$ (related to the establishment of a ferrimagnetic ground state) for the structure of low-energy excitations are dictated by the nonrelativistic version of Goldstone's theorem¹⁰: in the absence of long-range forces, a spontaneous symmetry breaking leads to low-energy excitations whose energy tends to zero for wave-vectors $k \rightarrow 0$. In contrast to the relativistic version, the theorem does neither specify the exact form of the dispersion relation for small k , nor does it determine the number of different Goldstone modes: these features are not fixed by symmetry considerations alone – rather, they depend on the specific properties of the system.

Turning to the case of Heisenberg ferromagnets, in a hydrodynamic description the quadratic form of the magnon energies

$$E_k = \frac{\rho_s}{M_0} k^2 + \mathcal{O}(k^4) \quad (2)$$

results entirely from the symmetry of the ferromagnetic state and the fact that the order parameter is itself a constant of the motion.¹¹ Here M_0 and ρ_s are, respectively, the magnetization density and the spin-stiffness constant of the Heisenberg ferromagnet. This form of the Goldstone modes (which is just the Landau-Lifshitz formula) can rigorously be obtained by simple sum-rule arguments (see, e.g., Ref. 12). Similar arguments are applicable to Heisenberg ferrimagnets as well.^{13,14} An alternative approach, relying on the suggested conformal invariance of the model (1), also predicts the quadratic form of Eq. (2).¹⁵ Due to the doubling of elementary cell in the ferrimagnetic ground state, there appears a second spin-wave branch in the ferrimagnetic case (optical magnons) with a finite energy gap Δ at $k = 0$: $\Delta \sim M_0/\chi_{||}$, where $\chi_{||}$ is the magnetic susceptibility parallel to \mathbf{M} .¹⁶ At intermediate temperatures, it is the optical magnon branch which produces a number of specific thermodynamic properties characteristic especially for the ferrimagnetic systems.^{2,17–20}

The existence of a macroscopic magnetic ground state also opens an interesting and rare opportunity to apply the spin-wave theory (SWT) to the low-dimensional quantum spin system (1). Indeed, recently published analysis based on the linear spin-wave approximation^{17,21,22} qualitatively confirmed the

expected structure of low-energy excitations in Heisenberg ferrimagnets. Moreover, the second-order SWT was shown²³ to produce precise quantitative results for the ground-state energy E_0 and the on-site magnetizations $m_1 = M_A/N$ and $m_2 = M_B/N$ even for the extreme quantum system $(S_1, S_2) = (1, \frac{1}{2})$: the second-order SWT results differ by less than 0.017% for E_0 , and by less than 0.18% for m_1 , from the density matrix renormalization group (DMRG) estimates.¹⁷ In this respect, an interesting question is to what extent the spin-wave approach can effectively be used to describe the properties of the 1D model (1) at zero temperature. A purpose of the present paper is to demonstrate, through an explicit study of the magnon-dispersion perturbation series, that the spin-wave approach can produce precise quantitative results for the magnon dispersions as well.

The paper is organized as follows. In Section 2, using the Dayson-Maleev boson representation of spin operators, the original spin Hamiltonian (1) is transformed to an equivalent boson Hamiltonian. The choice of an appropriate zeroth-order quadratic Hamiltonian for the perturbation series is also discussed here. In Section 3 we study the second-order corrections for the magnon energies and the related corrections for the spin-stiffness constant and the optical magnon gap. Third-order self-energy diagrams and the respective corrections for the optical magnon gap are also considered here. In the last Section the results are summarized and a comparison with available numerical estimates is made.

II. DYSON-MALEEV FORMALISM

To develop a spin-wave theory (see, e.g., Refs. 24,25 and references therein), one firstly transforms the original spin Hamiltonian (1) to an equivalent boson Hamiltonian. We adopt the following Dyson-Maleev representation of spin operators:

$$S_1^+(i) = \sqrt{2S_1} \left(a_i - \frac{1}{2S_1} a_i^\dagger a_i a_i \right), \quad (3)$$

$$S_1^-(i) = \sqrt{2S_1} a_i^\dagger, \quad S_1^z(i) = S_1 - a_i^\dagger a_i, \quad (4)$$

$$S_2^+(j) = \sqrt{2S_2} \left(b_j^\dagger - \frac{1}{2S_2} b_j^\dagger b_j^\dagger b_j \right), \quad (5)$$

$$S_2^-(j) = \sqrt{2S_2} b_j, \quad S_2^z(j) = b_j^\dagger b_j - S_2, \quad (6)$$

where $S_\alpha^\pm = S_\alpha^x \pm iS_\alpha^y$, $\alpha = 1, 2$. a_i and b_j are boson operators defined on the lattice sites $i \in \mathcal{A}$ and $j \in \mathcal{B}$, respectively.

Substituting the latter expressions in Eq. (1), one finds the following boson representation of \mathcal{H} in terms of the Fourier transforms a_k and b_k of a_i and b_j :

$$\mathcal{H}_B = -2\sigma S^2 N J + \mathcal{H}'_0 + V'_{DM}, \quad (7)$$

where

$$\mathcal{H}'_0 = 2SJ \sum_k \left[a_k^\dagger a_k + \sigma b_k^\dagger b_k + \sqrt{\sigma} \gamma_k \left(a_k^\dagger b_k^\dagger + a_k b_k \right) \right], \quad (8)$$

and

$$V'_{DM} = -\frac{J}{N} \sum_{1-4} \delta_{12}^{34} \times \quad (9)$$

$$(2\gamma_{1-4} a_3^\dagger a_2 b_1^\dagger b_4 + \sqrt{\sigma} \gamma_{1+2-4} a_3^\dagger b_2^\dagger b_1^\dagger b_4 + \frac{1}{\sqrt{\sigma}} \gamma_4 a_3^\dagger a_2 a_1 b_4).$$

$\gamma_k = \cos(ka_0)$ is the lattice structure factor and $\delta_{12}^{34} \equiv \delta(1+2-3-4)$ is the Kronecker δ -function. We have used the convention $(k_1, k_2, k_3, k_4) \equiv (1, 2, 3, 4)$ and the notations $\sigma \equiv S_1/S_2 > 1$ and $S_2 \equiv S$. Here and in what follows the external magnetic field is $H = 0$.

In the above expressions $\mathcal{H}'_0 = \mathcal{O}(S)$ is the quadratic boson Hamiltonian of the linear spin-wave theory (LSWT) and $V'_{DM} = \mathcal{O}(1)$ is the quartic Dyson-Maleev boson interaction. Omitting completely the boson interaction V'_{DM} , a number of authors has recently used the quadratic LSWT Hamiltonian \mathcal{H}'_0 for a qualitative description of the 1D ferrimagnetic model (1) at zero temperature.^{17,21,22} \mathcal{H}'_0 is easily diagonalized by use of the following Bogoliubov transformation to the quasiparticle boson operators α_k and β_k :

$$a_k = u_k(\alpha_k - x_k \beta_k^\dagger), \quad b_k = u_k(\beta_k - x_k \alpha_k^\dagger), \quad (10)$$

$$u_k = \sqrt{\frac{1 + \varepsilon_k}{2\varepsilon_k}}, \quad x_k = \frac{\eta_k}{1 + \varepsilon_k},$$

where $\varepsilon_k = \sqrt{1 - \eta_k^2}$ and $\eta_k = 2\sqrt{\sigma}\gamma_k/(1 + \sigma)$.

As a matter of fact, for the 1D ferrimagnetic model (1) the quadratic Hamiltonian (8) is not the most appropriate choice of a starting zeroth-order approximation for perturbation series. Since we are also interested in systems with small site spins, it is more appropriate from the very beginning to redefine \mathcal{H}'_0 by adding the first-order $1/S$ corrections to magnon dispersions.²⁶ The latter corrections come entirely from a normal ordering of the quasiparticle boson operators α_k and β_k in the quartic interaction V'_{DM} . It is important that these corrections (similar to Oguchi's corrections in antiferromagnets²⁷) renormalize the magnon excitation spectra (and the ground-state energy) without changing their basic structure, i.e., the number of Goldstone modes. Unlike the case of Heisenberg antiferromagnets, where Oguchi's corrections are numerically small even for site spins $S = \frac{1}{2}$, in quantum 1D ferrimagnets the magnon dispersions are significantly renormalized²⁶ (see below). As a result of the normal-ordering procedure, the boson Hamiltonian (7) is recast to the following basic form:

$$\mathcal{H}_B = E_0 + \mathcal{H}_0 + \lambda V, \quad V = V_2 + V_{DM}, \quad \lambda = 1. \quad (11)$$

The ground-state energy E_0 calculated up to first order in $1/S$ reads

$$\frac{E_0}{2NJ} = -\sigma S^2 + S [2\sqrt{\sigma} a_1 + (\sigma + 1) a_2] - a_1^2 - a_2^2 - \frac{\sigma + 1}{\sqrt{\sigma}} a_1 a_2, \quad (12)$$

where

$$a_1 = -\frac{\sqrt{\sigma}}{2(\sigma + 1)} \frac{1}{N} \sum_k \frac{\gamma_k^2}{\varepsilon_k}, \quad a_2 = -\frac{1}{2} + \frac{1}{2N} \sum_k \frac{1}{\varepsilon_k}. \quad (13)$$

Expressed in terms of quasiparticle operators, the corrected LSWT Hamiltonian reads

$$\mathcal{H}_0 = 2SJ \sum_k \left[\omega_k^{(\alpha)} \alpha_k^\dagger \alpha_k + \omega_k^{(\beta)} \beta_k^\dagger \beta_k \right], \quad (14)$$

where

$$\omega_k^{(\alpha, \beta)} = \left(1 - \frac{a_1}{S\sqrt{\sigma}} \right) \left(\frac{\sigma + 1}{2} \varepsilon_k \mp \frac{\sigma - 1}{2} \right) - a_2 \frac{1 - \gamma_k^2}{\varepsilon_k S}. \quad (15)$$

The normal-ordered quasiparticle interaction V contains a quadratic term,

$$V_2 = J \sum_k \left[V_k^{(+)} \alpha_k^\dagger \beta_k^\dagger + V_k^{(-)} \alpha_k \beta_k \right], \quad (16)$$

where

$$V_k^{(+, -)} = -\frac{a_2(\sigma - 1)^2}{\sqrt{\sigma}(\sigma + 1)} \left(1 \pm \frac{\sigma + 1}{\sigma - 1} \varepsilon_k \right) \frac{\gamma_k}{\varepsilon_k}, \quad (17)$$

and the quartic normal-ordered Dyson-Maleev interaction V_{DM} containing nine vertex functions: $V^{(i)} = V_{12;34}^{(i)}$, $i = 1, \dots, 9$. Explicit expressions for V_{DM} and the related vertex functions are presented in Appendix A.

The quadratic Hamiltonian (14) describes two branches of spin-wave excitations (magnons) defined with the dispersion relations $E_k^{(\alpha, \beta)} = 2SJ\omega_k^{(\alpha, \beta)}$, Eq. (15). The α excitations are gapless acoustical magnons in the subspace with $S_{tot}^z = (S_1 - S_2)N - 1$, whereas the β excitations are gapful optical magnons in the subspace $S_{tot}^z = (S_1 - S_2)N + 1$: $\mathbf{S}_{tot} = \sum_{n=1}^N [\mathbf{S}_1(n) + \mathbf{S}_2(n)]$. It is easy to show that the effect of the external magnetic field in Eq. (1) on the dispersions $E_k^{(\alpha, \beta)}$ reduces to the simple substitution $E_k^{(\alpha, \beta)} \mapsto E_k^{(\alpha, \beta)} \pm g\mu_B H$ provided that $g_1 = g_2 \equiv g$.

The spin stiffness constant ρ_s (related to the acoustical branch) plays a basic role (together with the magnetization density M_0) in the low-temperature thermodynamics of the model (1)^{6,18,28,29} and can be obtained from the Landau-Lifshitz relation (2) and Eq. (15):

$$\rho_s^{(0)} = Ja_0 S_1 S_2 \left[1 - \frac{a_1}{S\sqrt{\sigma}} - a_2 \frac{\sigma + 1}{S\sigma} \right]. \quad (18)$$

In the same free-quasiparticle approximation, the spectral gap of optical magnons at $k = 0$ reads²⁶

$$\Delta_0 = 2J(S_1 - S_2) \left(1 - \frac{a_1}{S\sqrt{\sigma}} \right). \quad (19)$$

In the case $(S_1, S_2) = (1, \frac{1}{2})$ Eqs. (18) and (19) give $\rho_s^{(0)}/Ja_0S_1S_2 = 0.761$ and $\Delta_0 = 1.676J$. On the other hand, the LSWT Hamiltonian \mathcal{H}'_0 produces the parameters of the related classical system: $\rho_s^{(0)}/Ja_0S_1S_2 = 1$ and $\Delta_0 = J$. The numerical estimate for the gap $\Delta = 1.759J$ ³⁰ clearly demonstrates the importance of the $1/S$ corrections in Eq. (15).

Next, let us consider the macroscopic sublattice magnetizations⁶ $m_1 = \langle S_1^z(i) \rangle = S_1 - \langle a_i^\dagger a_i \rangle$ and $m_2 = \langle S_2^z(j) \rangle = -S_2 + \langle b_j^\dagger b_j \rangle$ which are finite in the ferrimagnetic state. In the case of small site spins and for small magnetization densities M_0 , m_1 and m_2 are strongly reduced as compared to their classical values (respectively, S_1 and $-S_2$):

$$m_1 = S_1 - a_2, \quad m_1 + m_2 = S_1 - S_2. \quad (20)$$

As an example, for $(S_1, S_2) = (1, \frac{1}{2})$ Eq. (20) predicts a 42% reduction of the small spin $S_2 = \frac{1}{2}$. However, the off-diagonal quadratic interaction V_2 in Eq. (11) produces important first-order corrections for the sublattice magnetizations m_1 and m_2 . Thus, to first order in $1/S$ Eq. (20) should be replaced by the more precise expression

$$m_1 = S_1 - a_2 - \frac{\sqrt{\sigma}}{2S(\sigma+1)^2} \frac{1}{N} \sum_k (V_k^+ + V_k^-) \frac{\gamma_k}{\varepsilon_k^2}. \quad (21)$$

Now for the ferrimagnetic chain $(1, \frac{1}{2})$ the last expression gives the result $m_1 = 0.816$ which differs by only 3% from the DMRG numerical estimate 0.79248¹⁷.

Summarizing, it may be stated that the free-quasiparticle approximation based on the Hamiltonian (14) gives a good qualitative description of the ground-state properties of the model (1). Further improvement of the SWT results may be achieved by considering the role of quasiparticle interactions.

III. ROLE OF THE QUASIPARTICLE INTERACTIONS

Since the first-order $1/S$ corrections for the ground state energy E_0 and the dispersions $E_k^{(\alpha, \beta)}$ have already been taken into account by the normal-ordering procedure, corrections to Eqs. (12) and (15) arise only up from the second order of the perturbation series in V .

A. Second-order corrections for the magnon energies

The second-order corrections for the dispersion of acoustical magnons $\omega_k^{(\alpha)}$, Eq. (15), are connected with

the self-energy diagrams in Fig. 2. The respective analytic expressions read

$$\delta\omega_k^{(\alpha)}(a) = -\frac{1}{(2S)^2} \frac{V_k^{(+)} V_k^{(-)}}{\omega_k^{(\alpha)} + \omega_k^{(\beta)}}, \quad (22)$$

$$\delta\omega_k^{(\alpha)}(bc) = \frac{1}{(2S)^2} \frac{2}{N} \sum_p \frac{V_p^{(+)} V_{kp;pk}^{(2)} + V_p^{(-)} V_{kp;pk}^{(3)}}{\omega_p^{(\alpha)} + \omega_p^{(\beta)}}, \quad (23)$$

$$\delta\omega_k^{(\alpha)}(d) = -\frac{1}{(2S)^2} \frac{2}{N^2} \sum_{2-4} \delta_{k2}^{34} \frac{V_{43;2k}^{(8)} V_{k2;34}^{(7)}}{\omega_k^{(\alpha)} + \omega_2^{(\alpha)} + \omega_3^{(\beta)} + \omega_4^{(\beta)}}, \quad (24)$$

$$\delta\omega_k^{(\alpha)}(e) = -\frac{1}{(2S)^2} \frac{2}{N^2} \sum_{2-4} \delta_{k2}^{34} \frac{V_{43;2k}^{(3)} V_{k2;34}^{(2)}}{-\omega_k^{(\alpha)} + \omega_2^{(\beta)} + \omega_3^{(\alpha)} + \omega_4^{(\alpha)}}. \quad (25)$$

Note that since the vertex functions $V_k^{(-)}$, $V_{kp;pk}^{(2)}$, $V_{kp;pk}^{(3)}$, $V_{43;2k}^{(8)}$, and $V_{43;2k}^{(3)}$ vanish at the zone center $k = 0$ (see Appendix A), the gapless structure of $\omega_k^{(\alpha)}$ is preserved by each of the second-order corrections, Eqs. (22) to (25).

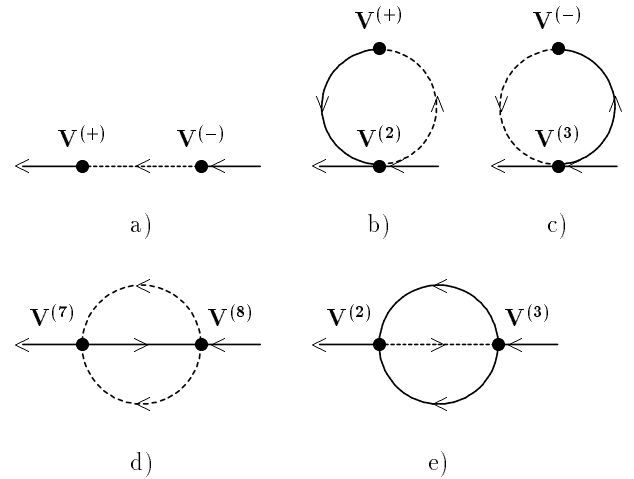


FIG. 2. Second-order self-energy diagrams giving the corrections for the dispersion of acoustical magnons $\omega_k^{(\alpha)}$. Solid and dashed lines represent, respectively, the propagators of α and β magnons. The Dyson-Maleev vertex functions $V^{(i)}$, $i = 1, \dots, 9$ are defined in Appendix A.

The second-order corrections for the dispersion of optical magnons $\omega_k^{(\beta)}$, Eq. (15), come from similar diagrams (see Fig. 3). The first diagram gives $\delta\omega_k^{(\beta)}(a) = \delta\omega_k^{(\alpha)}(a)$, whereas the other four diagrams give the following contributions:

$$\delta\omega_k^{(\beta)}(bc) = \frac{1}{(2S)^2} \frac{2}{N} \sum_p \frac{V_p^{(+)} V_{kp:pk}^{(5)} + V_p^{(-)} V_{kp:pk}^{(6)}}{\omega_p^{(\alpha)} + \omega_p^{(\beta)}}, \quad (26)$$

$$\delta\omega_k^{(\beta)}(d) = -\frac{1}{(2S)^2} \frac{2}{N^2} \sum_{2-4} \delta_{k2}^{34} \frac{V_{43;2k}^{(7)} V_{k2;34}^{(8)}}{\omega_k^{(\beta)} + \omega_2^{(\beta)} + \omega_3^{(\alpha)} + \omega_4^{(\alpha)}}, \quad (27)$$

$$\delta\omega_k^{(\beta)}(e) = -\frac{1}{(2S)^2} \frac{2}{N^2} \sum_{2-4} \delta_{k2}^{34} \frac{V_{43;2k}^{(5)} V_{k2;34}^{(6)}}{-\omega_k^{(\beta)} + \omega_2^{(\alpha)} + \omega_3^{(\beta)} + \omega_4^{(\beta)}}. \quad (28)$$

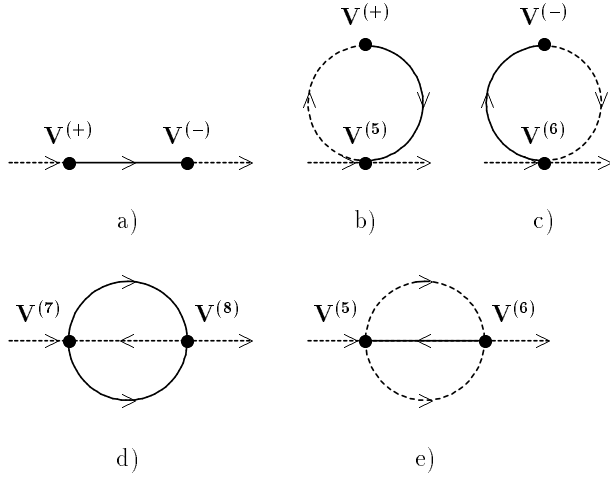


FIG. 3. Second-order self-energy diagrams giving the corrections for the dispersion of optical magnons $\omega_k^{(\beta)}$ (see also the notations in Fig. 2).

Note that in the present perturbation scheme (using \mathcal{H}_0 as a zeroth-order Hamiltonian), in Eqs. (22) to (28) there appear the renormalized dispersions $\omega_k^{(\alpha,\beta)}$. The standard $\mathcal{O}(1/S^2)$ corrections to the magnon dispersions can easily be obtained by substituting in Eqs. (22) to (28) the bare excitation energies (i.e., the functions $\omega_k^{(\alpha,\beta)}$ without the $1/S$ corrections). Since we are also interested in the extreme quantum systems which are composed of small site spins and have small magnetization densities M_0 [such as $(1, \frac{1}{2})$ and $(\frac{3}{2}, 1)$], it may be expected that the adopted perturbation scheme, where the quasiparticle interaction V is treated as a small perturbation, is more appropriate. Such a viewpoint is similar to

Oguchi's treatment of the Heisenberg antiferromagnet²⁷ and is supported by the following observations: (i) the higher-order corrections to the principle approximation for $\omega_k^{(\alpha,\beta)}$ are numerically small (see below), and (ii) the third-order series in V gives a somewhat better result for the gap Δ in the extreme quantum system $(1, \frac{1}{2})$. As a matter of fact, noticeable deviations from the standard $1/S$ expansions appear only in the above mentioned extreme quantum cases.

The dispersion functions $E_k^{(\alpha,\beta)}$ for the system $(1, \frac{1}{2})$, calculated up to second order in V , are presented in Fig. 4. For the optical magnon branch we find an excellent agreement with the exact-diagonalization results³⁰ in the whole Brillouin zone. Recently, a very successful description of the optical magnon branch of the system $(1, \frac{1}{2})$ has also been achieved through the variational matrix product approach.²⁰ As to the acoustical branch, the agreement with the available quantum Monte Carlo results³⁰ is not so satisfactory, especially near the zone boundary at $k = \pi/2a_0$. It is worth noticing that in the above region the calculated second-order corrections to $\omega_k^{(\alpha)}$, Eq. (15), are very small (about 0.5% of the principal approximation). In principle, it is not excluded that the spin-wave expansion partially fails to describe the acoustical branch in the system $(1, \frac{1}{2})$, as the latter is characterized by the minimal magnetization density $M_0 = 1/4a_0$ (see below). To clarify the problem, further studies are needed in this direction.

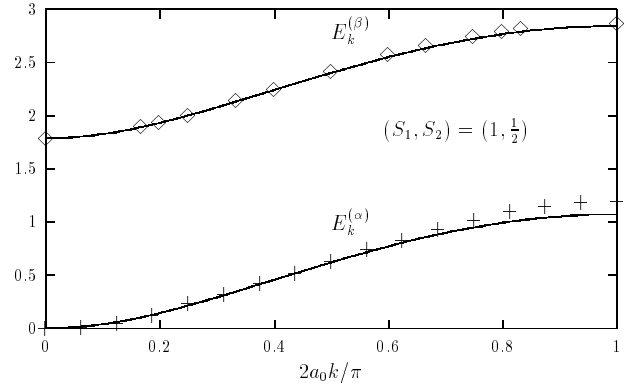


FIG. 4. Dispersions of the acoustical $E_k^{(\alpha)}$ and optical $E_k^{(\beta)}$ magnons (in J units) in the system $(S_1, S_2) = (1, \frac{1}{2})$ calculated up to second order in the magnon interaction V . The points (+ and \diamond) denote, respectively, quantum Monte Carlo and exact-diagonalization results (Ref. 30).

The above second-order corrections to $\omega_k^{(\alpha,\beta)}$ can be used to find the coefficient r_2 in the perturbation series for the spin-stiffness constant ρ_s

$$\frac{\rho_s}{Ja_0 S_1 S_2} = 1 + \frac{r_1}{2S} + \frac{r_2}{(2S)^2} + \mathcal{O}\left(\frac{1}{S^3}\right) \quad (29)$$

and the coefficient δ_2 in the series for the optical magnon gap

$$\frac{\Delta}{2J(S_1 - S_2)} = 1 + \frac{\delta_1}{2S} + \frac{\delta_2}{(2S)^2} + \frac{\delta_3}{(2S)^3} + \mathcal{O}\left(\frac{1}{S^4}\right). \quad (30)$$

Here $r_1 = -2a_1/\sqrt{\sigma} - 2a_2(\sigma + 1)/\sigma$ and $\delta_1 = -2a_1/\sqrt{\sigma}$ are obtained from Eqs. (18) and (19), respectively. The results for r_2 and δ_2 are presented in Table 1.

B. Third-order corrections for the optical magnon gap

The third-order corrections for the optical magnon gap Δ are connected with the self-energy diagrams in Fig. 5. These can be obtained by drawing all connected irreducible self-energy diagrams containing three vertex functions and only oppositely-oriented (α, β) pairs of internal magnon lines. The diagrams containing magnon lines closing on themselves vanish since the vertices have already been normal ordered. In addition, the diagrams which have internal α lines carrying the momentum k give zero contributions as well, since all vertex functions containing in-going α lines with momentum k vanish at $k = 0$ (see Appendix A).

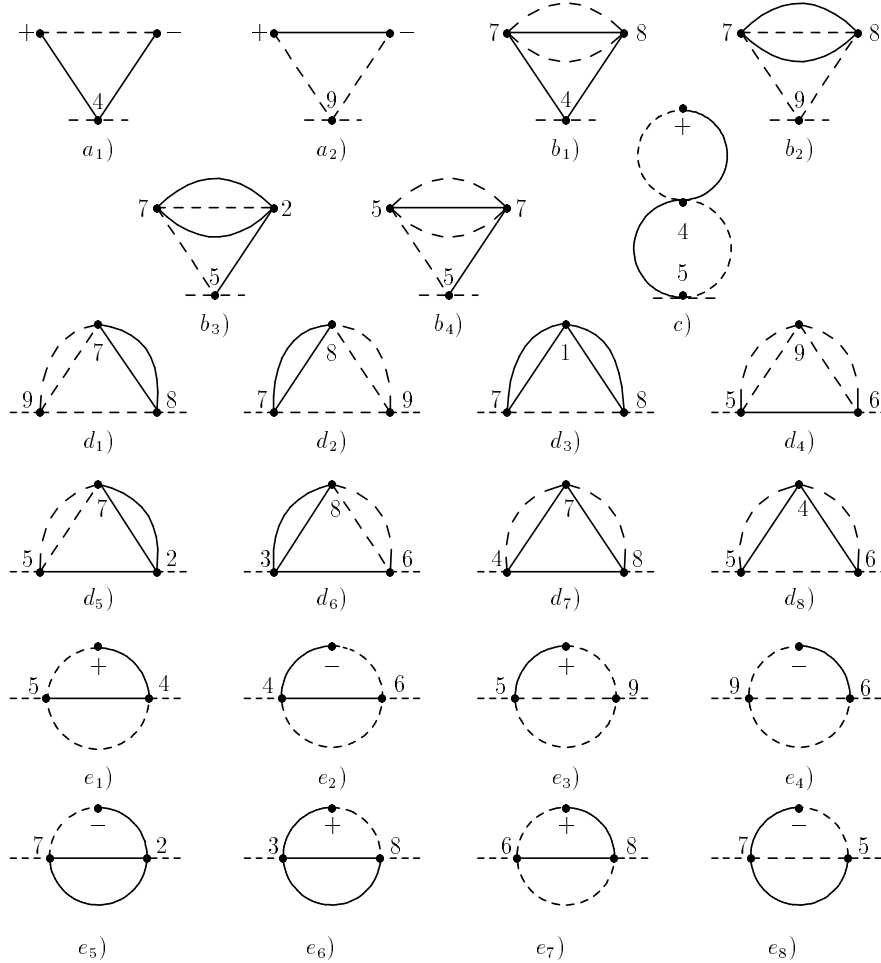


FIG. 5. Self-energy diagrams giving the third-order corrections for the optical magnon gap. The vertices are denoted by their superscripts. The diagrams b_1 , b_3 , c , d_7 , and d_8 represent groups of diagrams which can be obtained by the following vertex substitutions: $b_1)$ $(4,8,7) \mapsto (6,6,8)$; $b_3)$ $(5,2,7) \mapsto (6,3,8)$; $c)$ $(5,4,+)$ $\mapsto (6,4,-)$, $(6,8,+)$ (two diagrams), $(5,7,-)$ (two diagrams); $d_7)$ $(4,8,7) \mapsto (7,8,4)$, $(7,4,8)$; $d_8)$ $(5,6,4) \mapsto (6,6,8)$, $(5,5,7)$ (see also the notations in Fig. 2).

The analytic expressions related to the diagrams in Fig. 5 can easily be obtained by means of standard diagrammatic

rules (see, e.g., Ref. 31). For example, the diagrams a_1 , d_1 , and e_1 in Fig. 5 give the following contributions for the dispersion of optical magnons:

$$\delta\omega_k^{(\beta)}(a_1) = -\frac{1}{(2S)^3} \frac{2}{N} \sum_p \frac{V_p^{(+)} V_{pk;pk}^{(4)} V_p^{(-)}}{[\omega_p^{(\alpha)} + \omega_p^{(\beta)}]^2}, \quad (31)$$

$$\delta\omega_k^{(\beta)}(d_1) = -\frac{1}{(2S)^3} \frac{2}{N^3} \sum_{1-5} \delta_{12}^{34} \delta_{12}^{k5} \frac{V_{12;34}^{(7)} V_{34;5k}^{(9)} V_{k5;12}^{(8)}}{[\omega_1^{(\alpha)} + \omega_2^{(\alpha)} + \omega_3^{(\beta)} + \omega_4^{(\beta)}][\omega_1^{(\alpha)} + \omega_2^{(\alpha)} + \omega_k^{(\beta)} + \omega_5^{(\beta)}]}, \quad (32)$$

$$\delta\omega_k^{(\beta)}(e_1) = \frac{1}{(2S)^3} \frac{4}{N^2} \sum_{2-4} \delta_{k2}^{34} \frac{V_4^{(+)} V_{34;2k}^{(5)} V_{2k;43}^{(4)}}{[\omega_4^{(\alpha)} + \omega_4^{(\beta)}][-\omega_k^{(\beta)} + \omega_2^{(\alpha)} + \omega_3^{(\beta)} + \omega_5^{(\beta)}]}. \quad (33)$$

The results for δ_3 in systems with different site spins (S_1, S_2) are collected in Table 1. It is seen that the third-order corrections for Δ are numerically small (as compared to the second-order correction) even for the extreme quantum system $(S_1, S_2) = (1, \frac{1}{2})$.

IV. SUMMARY OF THE RESULTS AND DISCUSSION

The spin-wave results for the spin-stiffness constant ρ_s and the magnon gap Δ for a number of combinations

(S_1, S_2) are summarized in Table 1. The results for Δ are compared with available DMRG estimates.^{32,33} We find an excellent agreement with the numerical estimates³² for a number of systems, the largest deviation (about 0.88%) being connected with the system $(1, \frac{1}{2})$.

TABLE I. Spin-wave results for the spin stiffness constant ρ_s and optical magnon gap Δ of quantum Heisenberg ferrimagnetic chains containing two different spins $S_1 > S_2$ in the elementary cell. (r_1, r_2) and $(\delta_1, \delta_2, \delta_3)$ are, respectively, the coefficients of the spin-wave series for $r_s = \rho_s/Ja_0S_1S_2$, Eq. (29), and $\delta = \Delta/2(S_1 - S_2)$, Eq. (30). δ' indicates numerical estimates³² for the reduced excitation gap δ . $r_s^{(0)} = \rho_s^{(0)}/Ja_0S_1S_2$.

(S_1, S_2)	δ_1	δ_2	δ_3	δ	δ'	r_1	r_2	$r_s^{(0)}$	r_s
$(1, \frac{1}{2})$	0.6756	0.1095	-0.0107	1.7744	1.76	-0.2391	0.0283	0.7609	0.7892
$(\frac{3}{2}, 1)$	1.0428	0.4262	0.0812	1.6381	1.63	-0.4907	0.0184	0.7546	0.7592
$(\frac{3}{2}, \frac{1}{2})$	0.4013	0.0251	-0.0047	1.4217	1.42	-0.0959	0.0160	0.9041	0.9201
$(2, 1)$	0.6756	0.1279	-0.0103	1.3685	1.37	-0.2391	0.0391	0.8804	0.8902
$(2, \frac{1}{2})$	0.2861	0.0095	-0.0018	1.2938	1.29	-0.0517	0.0083	0.9483	0.9566

Since in the extreme quantum cases the constructed perturbation expansions basically rely on the suggested smallness of the quasiparticle interaction V , it is instructive to check the self-consistency of the theory by writing the series for ρ_s and Δ in terms of the formal parameter $\lambda = 1$, Eq. (11). Using the results from Table 1, the series for the system $(S_1, S_2) = (1, \frac{1}{2})$ read

$$\frac{\rho_s}{Ja_0S_1S_2} = 0.7609\lambda^0 + 0.0283\lambda^2 + \mathcal{O}(\lambda^3), \quad (34)$$

$$\frac{\Delta}{2J(S_1 - S_2)} = 1.6756\lambda^0 + 0.1095\lambda^2 - 0.0107\lambda^3 + \mathcal{O}(\lambda^4). \quad (35)$$

The above expansions explicitly demonstrate that the

quasiparticle interaction V in Eq. (11) introduces numerically small corrections to the zeroth-order principle approximation based on the quadratic quasiparticle Hamiltonian $\mathcal{H}_0 = \mathcal{O}(\lambda^0)$, Eq. (14). Of course, the smallness of corrections by itself does not ensure a good quality of the spin-wave expansion. The main weakness of the spin-wave theory is connected with the assumption that the long-range order is well established: it includes only transverse spin fluctuations, whereas the longitudinal spin fluctuations are completely neglected. In this respect, a typical example arises when spin-wave expansions are used for magnetic systems near the order-disorder critical point where the transverse and longitudinal fluctuations should be treated on equal ground. In spite of the fact that the corrections are small, the spin-wave series give unsatisfactory quantitative results.³⁴ On

the other hand, as the distance from the critical point increases the spin-wave description becomes more and more reliable. The indicated discrepancy for the acoustical branch in the extreme quantum system $(1, \frac{1}{2})$ (although the second-order corrections to the principle approximation are numerically small) probably reflects the discussed weakness of the SWT. As a matter of fact, taking the parameter M_0 as a measure of the distance from the disordered phase, such a behavior of the spin-wave series in the (S_1, S_2) family of 1D quantum Heisenberg ferrimagnets can also be indicated for the magnon gap Δ (see Table 1) and the parameters E_0 and m_1 (see Ref. 23): the largest deviations from the DMRG results appear in the cases with minimal magnetization density, $M_0 = 1/4a_0$. However, even in the extreme quantum system $(1, \frac{1}{2})$ the discrepancies are small and the spin-wave

expansion produces precise quantitative results. Concerning the dispersion of acoustical branch $\omega_k^{(\alpha)}$ (and the related spin-stiffness constant ρ_s), more numerical results are needed to make a statement about the accuracy of spin-wave description in this case. We believe, however, that at least for the systems with $M_0 > 1/4a_0$ the reported results for ρ_s closely approximate the true spin-stiffness constants at zero temperature.

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APPENDIX A: DYSON-MALEEV VERTICES FOR THE FERRIMAGNETIC MODEL

Using the symmetric form adopted in Ref. 35, the normal-ordered Dyson-Maleev quasiparticle interaction V_{DM} , Eq. (11), reads

$$\begin{aligned} V_{DM} = & -\frac{J}{2N} \sum_{1-4} \delta_{12}^{34} \left[V_{12;34}^{(1)} \alpha_1^\dagger \alpha_2^\dagger \alpha_3 \alpha_4 + 2V_{12;34}^{(2)} \alpha_1^\dagger \beta_2 \alpha_3 \alpha_4 + 2V_{12;34}^{(3)} \alpha_1^\dagger \alpha_2^\dagger \beta_3 \alpha_4 \right. \\ & + 4V_{12;34}^{(4)} \alpha_1^\dagger \alpha_3 \beta_4^\dagger \beta_2 + 2V_{12;34}^{(5)} \beta_4^\dagger \alpha_3 \beta_2 \beta_1 + 2V_{12;34}^{(6)} \beta_4^\dagger \beta_3^\dagger \alpha_2^\dagger \beta_1 \\ & \left. + V_{12;34}^{(7)} \alpha_1^\dagger \alpha_2^\dagger \beta_3^\dagger \beta_4^\dagger + V_{12;34}^{(8)} \beta_1 \beta_2 \alpha_3 \alpha_4 + V_{12;34}^{(9)} \beta_4^\dagger \beta_3^\dagger \beta_2 \beta_1 \right]. \end{aligned} \quad (A1)$$

The explicit form of the symmetric vertex functions for the ferrimagnetic Heisenberg model (1) is

$$V_{12;34}^{(i)} = u_1 u_2 u_3 u_4 \bar{V}_{12;34}^{(i)}, \quad i = 1, 2, \dots, 9, \quad (A2)$$

where

$$\begin{aligned} \bar{V}_{12;34}^{(1)} = & +\gamma_{1-3}x_1x_3 + \gamma_{1-4}x_1x_4 + \gamma_{2-3}x_2x_3 + \gamma_{2-4}x_2x_4 \\ & -\sigma^{1/2}(\gamma_{1-3-4}x_1x_3x_4 + \gamma_{2-3-4}x_2x_3x_4) - \sigma^{-1/2}(\gamma_1x_1 + \gamma_2x_2), \\ \bar{V}_{12;34}^{(2)} = & -\gamma_{1-3}x_1x_2x_3 - \gamma_{1-4}x_1x_2x_4 - \gamma_{2-3}x_3 - \gamma_{2-4}x_4 \\ & +\sigma^{1/2}(\gamma_{1-3-4}x_1x_2x_3x_4 + \gamma_{2-3-4}x_3x_4) + \sigma^{-1/2}(\gamma_1x_1x_2 + \gamma_2), \\ \bar{V}_{12;34}^{(3)} = & -\gamma_{1-3}x_1 - \gamma_{1-4}x_1x_3x_4 - \gamma_{2-3}x_2 - \gamma_{2-4}x_2x_3x_4 \\ & +\sigma^{1/2}(\gamma_{1-3-4}x_1x_4 + \gamma_{2-3-4}x_2x_4) + \sigma^{-1/2}(\gamma_1x_1x_3 + \gamma_2x_2x_3), \\ \bar{V}_{12;34}^{(4)} = & +\gamma_{1-3}x_1x_2x_3x_4 + \gamma_{1-4}x_1x_2 + \gamma_{2-3}x_3x_4 + \gamma_{2-4} \\ & -\sigma^{1/2}(\gamma_{1-3-4}x_1x_2x_3 + \gamma_{2-3-4}x_3) - \sigma^{-1/2}(\gamma_1x_1x_2x_4 + \gamma_2x_4), \\ \bar{V}_{12;34}^{(5)} = & -\gamma_{1-3}x_2x_3x_4 - \gamma_{1-4}x_2 - \gamma_{2-3}x_1x_3x_4 - \gamma_{2-4}x_1 \\ & +\sigma^{1/2}(\gamma_{1-3-4}x_2x_3 + \gamma_{2-3-4}x_1x_3) + \sigma^{-1/2}(\gamma_1x_2x_4 + \gamma_2x_1x_4), \\ \bar{V}_{12;34}^{(6)} = & -\gamma_{1-3}x_4 - \gamma_{1-4}x_3 - \gamma_{2-3}x_1x_2x_4 - \gamma_{2-4}x_1x_2x_3 \\ & +\sigma^{1/2}(\gamma_{1-3-4} + \gamma_{2-3-4}x_1x_2) + \sigma^{-1/2}(\gamma_1x_3x_4 + \gamma_2x_1x_2x_3x_4), \\ \bar{V}_{12;34}^{(7)} = & +\gamma_{1-3}x_1x_4 + \gamma_{1-4}x_1x_3 + \gamma_{2-3}x_2x_4 + \gamma_{2-4}x_2x_3, \\ & -\sigma^{1/2}(\gamma_{1-3-4}x_1 + \gamma_{2-3-4}x_2) - \sigma^{-1/2}(\gamma_1x_1x_3x_4 + \gamma_2x_2x_3x_4), \\ \bar{V}_{12;34}^{(8)} = & +\gamma_{1-3}x_2x_3 + \gamma_{1-4}x_2x_4 + \gamma_{2-3}x_1x_3 + \gamma_{2-4}x_1x_4 \end{aligned}$$

$$\begin{aligned}
& -\sigma^{1/2}(\gamma_{1-3-4}x_2x_3x_4 + \gamma_{2-3-4}x_1x_3x_4) - \sigma^{-1/2}(\gamma_1x_2 + \gamma_2x_1), \\
\bar{V}_{12;34}^{(9)} = & +\gamma_{1-3}x_2x_4 + \gamma_{1-4}x_2x_3 + \gamma_{2-3}x_1x_4 + \gamma_{2-4}x_1x_3 \\
& -\sigma^{1/2}(\gamma_{1-3-4}x_2 + \gamma_{2-3-4}x_1) - \sigma^{-1/2}(\gamma_1x_2x_3x_4 + \gamma_2x_1x_3x_4).
\end{aligned}$$

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